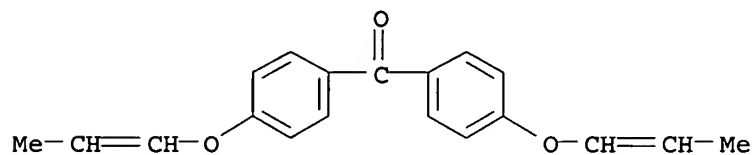


10/690,527

ANSWER 16 OF 36 CAPLUS COPYRIGHT 2006 ACS on STN

AN 1993:540479 CAPLUS
DN 119:140479
TI Heat-resistant resin compositions for sealing semiconductors
IN Kanayama, Kaoru; Ichikawa, Shuji
PA Mitsubishi Petrochemical Co., Ltd., Japan
SO Jpn. Kokai Tokkyo Koho, 9 pp.
CODEN: JKXXAF
DT Patent
LA Japanese
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	PATENT NO.	KIND	DATE	APPLICATION NO.	DATE
PI	JP 05051418	A2	19930302	JP 1991-236795	19910826 <--
PRAI	JP 1991-236795		19910826		
IT	149829-41-4P				
	RL: PREP (Preparation) (preparation of)				
RN	149829-41-4 CAPLUS				
CN	Methanone, bis[4-(1-propenyloxy)phenyl]- (9CI) (CA INDEX NAME)				



$$p = 1$$

$$m = 0$$

$$r = 1$$

$$w = c = 0$$

$$q = 1$$

$$n = 0$$

$$R = R' = (3)$$

$$R^a = R^b = H$$

$$R^c = Me$$

102 -7 1

PATENT ABSTRACTS OF JAPAN

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(71)Applicant : MITSUBISHI PETROCHEM CO
LTD

(22)Date of filing : 26.08.1991

(72)Inventor : KANAYAMA KAORU
ICHIKAWA SHUJI

(54) SEALING RESIN COMPOSITION

(57)Abstract:

PURPOSE: The subject composition comprising a poly unsaturated dicarboxylic acid imide compound, 1-propenyl etherified aromatic compound, a polymerization catalyst and an inorganic filler in specific amounts, having excellent moldability and heat resistance, and giving cured products reduced in the water-absorbing property.

CONSTITUTION: The objective composition comprising (A) a poly unsaturated dicarboxylic acid imide compound (e.g. N,N'-ethylene bismaleimide) having two or more unsaturated dicarboxylic imide groups in the molecule, (B) a 1- propenyl etherified aromatic compound having two or more 1-propenyloxy groups in the molecule, (C) a polymerization catalyst (e.g. benzoyl peroxide), and (D) an inorganic filler (e.g. silica fine particles) in an A/B weight ratio of 0.05-20.0, a C/(A+B) weight ratio of 0.001-0.01, and a (A+B+C)/D weight ratio of 0.1-10.

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3. In the drawings, any words are not translated.

CLAIMS

[Claim(s)]

[Claim 1] Component : (A) The Pori partial saturation dicarboxylic acid imide compound which has at least two partial saturation dicarboxylic acid imide radicals in 1 molecule, (B) -- component: -- 1-propenyl etherification aromatic series system compound which has at least two 1-propenyloxy radicals in 1 molecule -- It comes to contain each component of a polymerization catalyst and a (D) component: inorganic bulking agent. (C) -- component: -- (-- A --) -- a component -- /-- (-- B --) -- a component -- weight -- a ratio -- 0.05 - 20.0 -- (-- C --) -- a component -- /-- [-- (-- A --) -- a component -- + -- (-- B --) -- a component --] -- weight -- a ratio -- 0.001 - zero . -- one -- [-- (-- A --) -- a component -- + -- (-- B --) -- a component -- + -- (-- C --) -- a component --] -- /-- (-- D --) -- a component -- weight -- a ratio -- 0.1 - ten -- it is -- things -- the description -- ** -- carrying out -- closure -- ** -- resin -- a constituent .

[Claim 2] (A) The resin constituent for closure according to claim 1 whose Pori partial saturation dicarboxylic acid imide compound of a component is a polymer lane acid imide compound.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention is excellent in fabricating-operation nature useful as the resin constituent for closure, especially an object for semi-conductor closure, and relates to the resin constituent for closure which is moreover excellent in thermal resistance, and gives a hardened material with little absorptivity.

[0002]

[Description of the Prior Art] Conventionally, electronic parts, such as diode, a transistor, and an integrated circuit, are closed using thermosetting resin. Since it is advantageous in respect of economical and others many compared with hermetic sealing which used glass, a metal, the ceramics, etc., this resin seal is put in practical use widely.

[0003] As a resin constituent for this kind of resin seals, the epoxy resin constituent has mainly been used in respect of dependability, a price, etc. also in much thermosetting resin. However, it is in the inclination for the service condition of closure electronic parts to become severe gradually in recent years, and the demand to the thermal resistance of a closure resin ingredient is becoming severe especially.

[0004] Moreover, although thermosetting polymaleimide resin constituents, such as a bismaleimide resin (the European Patent No. 1770867 description) containing aromatic series diamine and resin (JP,55-39242,B) which consists of an allyl compound phenol which has two aromatic series nuclei, and bismaleimide, are conventionally known as high heat resistant resin, these need the heating at high temperature of long duration at the time of a fabricating operation, and have a fault with the large water absorption of a resin hardened material.

[0005]

[Problem(s) to be Solved by the Invention] This invention is excellent in fabricating-operation nature, and tends to offer the resin constituent for closure which is moreover excellent in thermal resistance, and can give hardening resin with low water absorption.

[0006]

[Means for Solving the Problem] The Pori partial saturation dicarboxylic acid imide compound with which the resin constituent for closure of this invention has at least two partial saturation dicarboxylic acid imide radicals in (A) component: 1 molecule, (B) -- component: -- 1-propenyl etherification aromatic series system compound which has at least two 1-propenyloxy radicals in 1 molecule -- It comes to contain each component of a polymerization catalyst and a (D) component: inorganic bulking agent. (C) -- component: -- (-- A --) -- a component -- /-- (-- B --) -- a component -- weight -- a ratio -- 0.05 - 20.0 -- (-- C --) -- a component -- /-- [-- (-- A --) -- a component -- + -- (-- B --) -- a component --] -- weight -- a ratio -- 0.001 - zero . -- one -- [-- (-- A --) -- a component -- + -- (-- B --) -- a component -- + -- (-- C --) -- a component --] -- /-- (-- D --) -- a component -- weight -- a ratio -- 0.1 --- ten -- it is - things -- the description -- ** -- carrying out -- a constituent -- it is .

[0007] As a Pori partial saturation dicarboxylic acid imide compound used as a (A) component in this

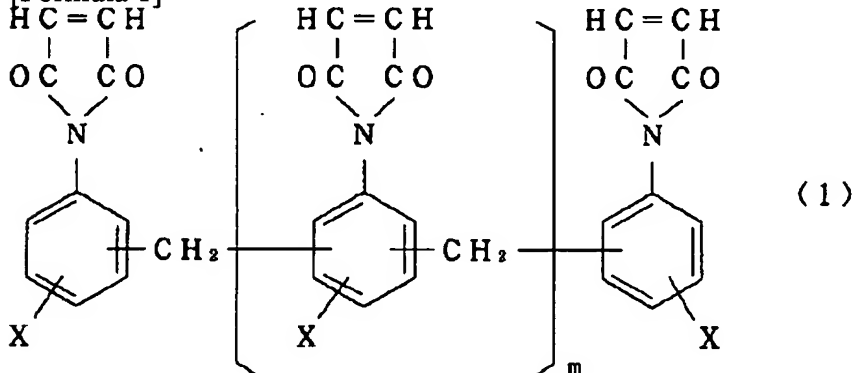
invention, the compound of a publication is raised to following ** - **.

** N and N'-ethylene bismaleimide, N, and N'-hexamethylene bismaleimide, N and N'-m-phenylene bismaleimide, N, and N'-p-phenylene bismaleimide, The N, N'-4, and 4'-diphenylmethane bismaleimide, N, N'-3, and 3'-dimethyl -5, 5'-diethyl-diphenylmethane bismaleimide, N, N'-4, and 4'-diphenyl ether bismaleimide, N,N'-methylenebis (3-chloro-p-phenylene) bismaleimide, N, N'-4, and 4'-diphenylsulfone bismaleimide, N, N'-4, and 4'-dicyclohexyl methane bismaleimide, N and N'-alpha, alpha'-4, and 4'-dimethylene cyclohexane bismaleimide, N and N'-meta xylene bismaleimide, N, N'-4, and 4'-diphenyl cyclohexane bismaleimide, The N, N'-4, and 4'-triphenyl -1, 1-propane bismaleimide, N, N'-4, and 4'-triphenyl -1, 1, and 1-ethane bismaleimide, N, N'-4, and 4'-triphenylmethane color bismaleimide, N, N'-3, 5-triazole -1, 2, 4-bismaleimide, Bismaleimide, such as a 2 and 2-bis[4-(4-maleimide phenoxy)-phenyl] propane and a bis[4-(maleimide phenoxy) phenyl] sulfone.

[0008] ** Polymaleimide expressed with the following general formula (1).

[0009]

[Formula 1]

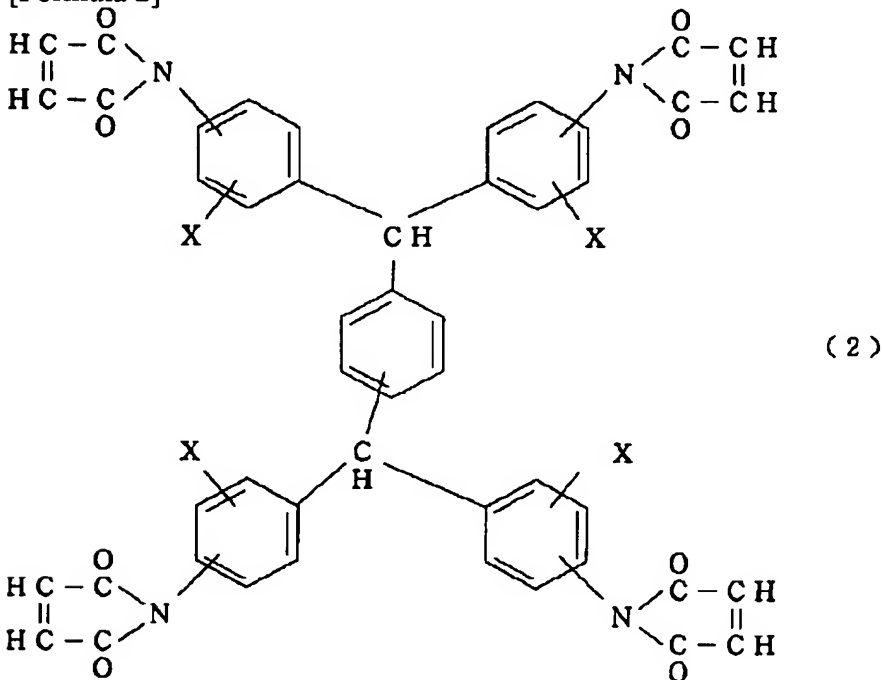


[0010] The alkyl group of a hydrogen atom, a halogen atom, or carbon numbers 1-4 or this alkoxy group, and m of X are the integers of 0-4 among this formula.

[0011] ** Polymaleimide expressed with the following general formula (2) (in detail refer to JP,59-12931,A).

[0012]

[Formula 2]



[0013] X is the alkyl group or this alkoxy group of a hydrogen atom, a halogen atom, or carbon numbers 1-4 among this formula.

[0014] ** Mixture of the polymaleimide expressed with the aforementioned general formula (1), and the polymaleimide expressed with the aforementioned general formula (2) (refer to JP,60-26032,A).

[0015] ** N and N'-m-phenylene-screw-SHITORAKONIMIDO, N, N'-4, and 4'-diphenylmethane-screw-SHITORAKONIMIDO, N, N'-4, and 4'-diphenylmethane-screw-itaconimide, N, and N'-p-phenylene-screw-itaconimide, N, N'-4, and 4'-diphenylmethane-screw-dimethyl maleimide, N, N'-4, 4'-2, and 2'-diphenyl propane-screw-dimethyl maleimide, N and N'-hexamethylene-screw-dimethyl maleimide, N, N'-4, 4'-(diphenyl ether)-Screw-dimethyl maleimide and N, N'-4, and 4'-diphenylsulfone-screw-dimethyl maleimide etc.

[0016] In this invention, the Pori partial saturation dicarboxylic acid imide which has these Pori partial saturation dicarboxylic acid imide radicals can also use two or more sorts together.

[0017] 1-propenyl etherification aromatic series system compound used as a (B) component in this invention can be easily manufactured by processing a corresponding allyl compound etherification aromatic series system compound under existence of a divalent palladium compound catalyst, and isomerizing the end double bond of an allyl compound ether group to an internal double bond.

[0018] The allyl compound etherification aromatic series system compound of the raw material can be easily manufactured by making the aromatic series system compound and allyl compound halide (for example, an allyl chloride, an allyl compound star's picture, etc.) which have at least two phenolic hydroxyl groups react under existence of bases, such as a sodium carbonate, and an organic solvent [for example, refer to a journal OBU American chemical society and the 1863rd volume [62nd] page (1940)].

[0019] furthermore, as an aromatic series system compound which has at least two phenolic hydroxyl groups used as the raw material for manufacturing the allyl compound etherification aromatic series system compound A catechol, hydroquinone, biphenol, 2, and 2-bis(4-hydroxyphenyl) propane [namely, bisphenol A], Bis(4-hydroxyphenyl) methane [namely, the bisphenol F], A 4 and 4-dihydroxy benzophenone, 4, and 4'-dihydroxy phenyl sulfone, 3, the 9-screws (2-hydroxyphenyl) 2, 4, and 8, a 10-tetraoxaspiro [5 and 5]-undecane, 3, 9-screw (4-hydroxyphenyl) - 2, 4, 8, a 10-tetraoxaspiro [5 and 5]-undecane, 1, 1, 1, 3, 3, and 3-hexafluoro - Dihydric phenols, such as a 2 and 2-bis(p-hydroxyphenyl) propane [namely, hexafluoro bisphenol A] The polyphenol which a phenol novolak, a cresol novolak, salicylaldehyde and a phenol, or cresol is made to react under an acid catalyst, and is obtained The polyphenol which a p-hydroxy benzaldehyde, a phenol, or cresol is made to react under an acid catalyst, and is obtained Polyhydric phenols, such as polyphenol which a terephthal aldehyde, a phenol, cresol, or a bromine phenol is made to react under an acid catalyst, and is obtained, are raised.

[0020] In this invention, one kind may be used as a (B) component, two or more sorts are used together, and 1-propenyl etherification aromatic series system compound manufactured as mentioned above can also be considered as the (B) component.

[0021] the blending ratio of coal of the Pori partial saturation dicarboxylic acid imide compound (A) in this invention, and 1-propenyl etherification aromatic series system compound (B) -- (A) component / (B) component weight ratio -- 0.05-20.0 -- it is 0.25-5.0 preferably. (A) It becomes the inclination for a hardened material to become weak if there is too much blending ratio of coal of a component, becomes the inclination for the thermal resistance of a hardened material to become low if there are too few rates of the (A) component, and is not all desirable.

[0022] As a polymerization catalyst of the (C) component in this invention, radical polymerization initiators, such as organic peroxide and an azo compound, are mentioned. When the radical polymerization initiator is illustrated, benzoyl peroxide, G t-butyl peroxide, JIKUMIRU peroxide, lauroyl peroxide, acetyl peroxide, Methyl-ethyl-ketone peroxide, cyclohexanone peroxide, t-butyl hydro peroxide, azobisisobutyronitril, 1, and 1'-azobis (cyclohexane-1-carbonitrile), 2 and 2'-azobis (2-amidinopropane) dihydrochloride, 2, and 2'-azobis [2-(2-imidazo RIIN-2-IRU) propane], 2, and 2'-azobis (2, 4, and 4-trimethyl pentane) etc. is raised. The class of polymerization catalyst to be used and

an addition can adjust a cure rate. Since JIKUMIRU peroxide, azobisisobutyronitril, 1, and 1'-azobis (cyclohexane-1-carbonitrile) etc. is 0.1 - 5% of the weight of addition and shaping hardening of it is attained to the most practical process condition of the closure ingredient which is the field of the invention of the resin constituent of this invention, i.e., the sum total weight of a (A) component + (B) component, with heating for bottom of temperature of about 175 degrees C 1 - 5 minutes, it is especially desirable.

[0023] this invention -- it can set -- (-- C --) -- a component -- namely, -- a polymerization catalyst -- the blending ratio of coal -- (-- C --) -- a component -- /-- [-- (-- A --) -- a component -- + -- (-- B --) -- a component --] -- weight -- a ratio -- 0.001 to 0.1 -- it is 0.005-0.05 preferably. (C) Since the electrical property of a hardened material etc. will deteriorate if hardening at the time of a fabricating operation will become inadequate if there are too few rates of a component, and there are too many the rates, neither is desirable.

[0024] As an inorganic bulking agent (D) in this invention, an impalpable powder silica, fused silica, a crystalline silica, an aluminum hydroxide, an aluminum oxide, magnesium oxide, a mica, talc, a titanium white, a calcium carbonate, a glass fiber, etc. are raised, for example. These are independent or are used as two or more sorts of mixed stock. A powdered silica, fused silica, and especially a crystalline silica are desirable also in these.

[0025] The range of the blending ratio of coal of the (D) component in this invention is 0.1-10 in a [(A) component + (B) component + (C) component] / (D) component weight ratio. (D) If too few [if the rate of a component increases too much, the fluidity of resin will be lost and fabricating-operation nature will worsen, and], since the mechanical characteristic at the time of the elevated temperature of a hardened material will fall, neither of the cases is desirable.

[0026] To the resin constituent of this invention, a flame retarder, a coloring agent, a pigment, etc. can be added if needed (as an arbitration component). As the flame retarder, organic ****, such as inorganic ****, such as *****, and triphenyl phosphate, etc. are raised. As the coloring agent thru/or a pigment, a titanium dioxide, the chrome yellow, carbon black, iron black, a molybdate orange, etc. are raised, for example. The blending ratio of coal of these arbitration component has less than 50 desirable % of the weight of the amount of the range which does not spoil the original property of the resin constituent of this invention, i.e., all resin.

[0027] Moreover, it is also possible to add the epoxy resin which reacts to the resin constituent of this invention with a phenolic hydroxyl group if needed, and gives the structure of cross linkage.

[0028] Adjustment of the resin constituent of this invention is performed by if possible mixing to homogeneity each component and the arbitration component which can be blended further if needed of (A) - (D) by mixing suitably, i.e., means, such as kneading using heating melting mixing, a roll, a kneader, etc., mixing using a suitable organic solvent, and dry blending.

[0029]

[Example] Below, the synthetic example (this is only called "synthetic example" below) example and the example of a comparison of 1-propenyl etherification aromatic series system compound are given and explained in full detail. The "section" given in these examples means the weight section.

[0030] Bisphenol A (228g), NaOH (28.5g), and n-propanol 1000ml were taught to the 4 opening flask with a capacity of 2000ml which attached synthetic example 1 (allyl compound etherification process) thermometer, the agitator, the cooling pipe, and the dropping funnel, and contents were dissolved in it under heating reflux. After dissolving thoroughly, when 200ml of allyl chlorides was dropped gradually, as for after the completion of whole-quantity dropping, the system of reaction became neutrality. n-propanol was distilled off, after heating for 3 hours, making it react under reflux succeedingly and carrying out precipitation of NaCl cooled and generated the ** exception.

[0031] It dissolved in the methylene chloride, 308g of diaryl ether ghosts of obtained rough bisphenol A was rinsed, and the methylene chloride was distilled out of the oil reservoir after separating a water layer. The structure of the obtained purification bisphenol A diaryl ether was identified by liquid chromatographic analysis, nuclear magnetic resonance analysis, and infrared-absorption spectral analysis.

[0032] (1-propenyl etherification process) Teaching purification bisphenol A diaryl ether 103g (0.33 mols) obtained at the aforementioned process, 200g of carbon tetrachlorides, and dichlorobis (acetonitrile) palladium (II) 7.76g (0.02 mols) to the 4 opening flask which attached stirring equipment, a cooling pipe, a thermometer, and gas installation tubing, and warming it at 50 degrees C with an oil bath under nitrogen-gas-atmosphere mind, it stirred for 8 hours and the isomerization of 1-propenyl etherification was made to perform.

[0033] After moving reaction generation liquid to the evaporator, warming under reduced pressure and distilling off a carbon tetrachloride (recovery), 40ml of petroleum ether was added and stirred to the residue, and filtration under reduced pressure removed orange precipitation of dichlorobis (acetonitrile) palladium (II) (recovery). It distilled off by warming under reduced pressure of the filtrate (recovery), and 100g of transparent and colorless viscosity-like liquids was obtained. This liquid was identified the *****A** (1-propenyl) ether by each analysis of liquid chromatography, gel filtration chromatography, nuclear magnetic resonance, and infrared spectroscopy. The rate of isomerization was 100%.

[0034] After teaching phenol 940g, salicylaldehyde 122g, and 0.2g of sulfuric acids to a three-neck flask with a capacity of 2000ml which attached synthetic example 2 (polyphenol composition process) thermometer, the agitator, and the cooling pipe and carrying out temperature up of the internal temperature to it to 110 degrees C, it held to this temperature for further 4 hours, and was made to react to it. Methyl-isobutyl-ketone (it is hereafter called "MIBK" for short) 1000ml was added and diluted after reaction termination using the separating funnel, 600ml distilled water washed 3 times, and the sulfuric acid was removed. The obtained solution was moved to the rotary evaporator, it distilled off under MIBK and reduced pressure of a superfluous phenol, and dark reddish-brown glass-like polyphenol was obtained. This polyphenol was the melting point of 111 degrees C, and average molecular weight 353.

[0035] (Allyl compound etherification process) After stirring until it taught 1000ml of n-propyl alcohol, and NaOH 180g to the 4 opening flask with a capacity of 2000ml which attached the thermometer, the agitator, the condensator, and the dropping funnel and became homogeneity, polyphenol 450g obtained at the above-mentioned process was added, and it stirred for further 1 hour. Subsequently, after being dropped at it, covering [298g] it over this for 10 minutes, temperature up of the reaction mixture is carried out to 100 degrees C, and it stirred for 3 hours and was made to react at this temperature.

[0036] After removing the salt which carried out the resultant the ** exception and carried out the byproduction, it distilled off under reduced pressure of n-propyl alcohol (recovery), and 560g of liquid resultants with viscous dark reddish-brown was acquired. When this resultant performed each analysis of nuclear magnetic resonance and a gel permeation chromatograph, it was only an allyl compound etherification object of polyphenol.

[0037] (1-propenyl etherification process) Polyphenol allyl compound ether 560g (4.0 mols) obtained at the aforementioned process, chloroform 1490g (1000ml), and dichlorobis (acetonitrile) palladium (II) 2.86g (11 millimol) were taught to the same 4 opening flask also in the synthetic example 1, and it warmed at 46 degrees C, it stirred at this temperature for 6 hours, and the isomerization of 1-propenyl etherification was made to perform.

[0038] This reaction generation liquid was processed by the same approach as the synthetic example 1, and 550g of dark reddish-brown transparence viscosity-like liquids was obtained. When this generation liquid was analyzed by the same approach as the synthetic example 1, it was 1-propenyl etherification object of polyphenol, and the rate of isomerization from that allyl compound ether was 100%.

[0039] Using the raw material and reaction condition which are shown in three to synthetic example 5 table 1, others carry out allyl compound etherification using an approach and a reaction condition given in the synthetic example 1, isomerized the obtained allyl compound etherification object, and carried out 1-propenyl etherification. The name, the yield, and the rate of isomerization of obtained 1-propenyl etherification object were as being shown in a table 1.

[0040]

[A table 1]

表 1

例	アリルエーテル化工工程			1-プロペニルエーテル化工工程						
	原 材 料		塩基 (部)	原 材 料		反応条件		1-プロペニルエーテル化合物		
	芳香族系化合物 (部)	アリルハ ライド (部)		ア リ ル エ ー テ ル 化 物 (部)	触媒 (部)	温度 (℃)	時間 (hr)	名 称	収量 (部)	異性化 率 (%)
合成例 3	4,4'-ジヒドロキシベンゾフェノン (214)	塩化アリ ル (154)	NaOH (80)	*1 (100)	ジクロロ ビス(ア セトニ トリル) パラジ ウム (II) (0.93)	50	5	4,4'-ビス(1-プロペニル)ベンゾフェノン	94	99
合成例 4	ビス(2-ヒドロキシフェニル)メタン(すなわちビスフェノールF) (200)	塩化アリ ル (154)	Na ₂ CO ₃ (212)	*1 (100)	(同上) (1.0)	55	6	ビス(1-プロペニル)エタニン	96	100
合成例 5	ビス(4-ヒドロキシフェニル)スルホン(すなわちビスフェノールS) (250)	臭化アリ ル (241)	NaOH (80)	*1 (100)	ジクロロ ビス(ベ ンゾニ トリル) パラジ ウム (II) (1.5)	63	4	ビス(4-プロペニル)エタニン	95	97

[0041] The notes of a table 1: *1 ... What was obtained with the allyl compound etherification process of each example was used for the raw material allyl compound etherification object of 1-propenyl etherification process.

[0042] After carrying out melting kneading for 10 minutes using the 6 inch roll which heated the diamino diphenylmethane bismaleimide (Mitsubishi Petrochemical Co., Ltd. trade name MB-3000) 57 section of example 1 marketing, the 1-propenyl etherification object 43 section compounded in the

synthetic example 1, and the thing which mixed the dicumyl peroxide (DCPO) 1.0 section and the fused silica 233 section as a polymerization catalyst at 100-130 degrees C, it ground after cooling and the powder for shaping was obtained. The gel time in 175 degrees C of this powder for shaping was 48 seconds. The account of ** of the above main point was carried out to the after-mentioned table 2.

[0043] Subsequently, after transfer-molding the obtained powder for shaping using the conditions for [175 degree-Cx70 kg/cm] 2x2 minutes, it heated at 200 degrees C for 5 hours, and postcure was carried out to them. The account of ** of the transfer-molding condition, hardened material physical properties, and the spiral flow value of the powder for shaping was carried out to the table 3 which carries out a postscript.

[0044] Others prepared various molding resin constituent powder according to the example 1 using the resin presentation shown in one to example 2 - example of 7 comparisons 2 table 2. The gel time in 175 degrees C of each obtained powder of resin each, was as being shown in a table 2.

[0045]

[A table 2]

表 2

例	樹脂組成物					ゲルタイム (175℃)
	樹	脂	樹脂組成成分	重合触媒	熔融シリカ	
	(部)					
	ポリマレイミド化合物					
1	ジアミノジフェニルメタンビスマレイミド*1	(57)	合成例1で合成した1-プロペニルエーテル化合物	DCPO*2 (1.0)	(233)	48 秒
2	同上	(56)	合成例2で合成した1-プロペニルエーテル化合物	同上 (2.0)	同上	40 秒
3	同上	(56)	同上	ACCN*3 (1.5)	同上	55 秒
4	2,2-ビス(4-(4-マレイミドフェニル)プロパン)プロパン*4	(67)	同上	DCPO*2 (1.0)	同上	43 秒
5	ジアミノジフェニルメタンビスマレイミド*1	(55)	合成例3で合成した1-プロペニルエーテル化合物	AIBN*5 (1.5)	同上	50 秒
6	同上	(56)	合成例4で合成した1-プロペニルエーテル化合物	ACCN*3 (2.0)	同上	52 秒
7	同上	(52)	合成例5で合成した1-プロペニルエーテル化合物	DCPO*2 (2.0)	同上	47 秒
1	同上	(54)	0,0'-ジアリルビスフェノールA	同上 (2.0)	同上	21 分
2	同上	(54)	ビスフェノールAジアリルエーテル	同上 (2.0)	同上	39 分

[0046] The notes of a table 2: *1 ... Mitsubishi Petrochemical Co., Ltd. trade name MB-3000*2 ... Abbreviated-name *3 of dicumyl peroxide ... 1 1' - Abbreviated-name *4 of azobis (cyclohexane-1-carbonitrile) ... Mitsubishi Petrochemical Co., Ltd. trade name MB-8000*5 ... Abbreviated name of azobisisobutyronitril [0047] Moreover, after fabricating using the transfer-molding conditions which measure the spiral flow value same in the example 1 about each shaping powder obtained in the above-mentioned examples 2-7 and the examples 1-2 of a comparison as eye a bucket, and are shown in a table

3, various kinds of physical-properties values of the hardened material which heated for 5 hours and carried out postcure at 200 degrees C were measured. The result was as being shown in a table 3.

[0048] It fabricated on the transfer-molding conditions which 690 ***** and others apply the 96 sections and triphenyl phosphine to the two sections correspondingly, apply [epoxy resin / (oil-ized shell epoxy incorporated company trade name Epicoat 180H) / of example of comparison 3 marketing / orthochromatic cresol novolak system] fused silica to the approach of an example 1 correspondingly in the 200 sections and a phenol novolak (Gunei Chemical Industry Co., Ltd. trade name PSF-4261), manufacture shaping powder, and show the obtained powder in a table 3, and postcure was carried out on the hardening conditions of 200 degree-Cx 5 hours. The spiral flow value of the shaping powder and various kinds of physical properties of a hardened material were as being shown in a table 3. Moreover, the gel time in 175 degrees C of this powder for shaping was 32 seconds.

[0049]

[A table 3]

表 3

例	トランスファー 成形条件		スパイ ラルフ ロー (cm)	硬 化 物 物 性						吸水率 #5 (%)		
				Tg #1 (°C)	α #2 [$\times 10^{-6}$ (1/°C)]	曲 げ 試 験 値 #3 (単位はいずれも Kg · f/mm ²)		Td #4 (5重量 %ロス) (°C)				
	2 3 °C								2 6 0 °C			
	温度 (°C)	時間 (min)				強度	弾性率		強度		弾性率	
実 施	1	175	2	75	254	1.48	10.7	2189	6.72	886	409	1.05
	2	175	2	70	272	1.32	11.7	1806	7.66	942	412	0.98
	3	175	2	79	278	1.34	11.0	1825	7.53	1000	410	1.00
例	4	175	2	73	269	1.32	11.5	1808	7.48	987	411	1.03
	5	175	2	77	260	1.40	11.3	1851	7.07	899	412	0.99
	6	175	2	77	244	1.50	10.1	1857	5.8	854	405	1.24
比 較 例	7	175	2	75	270	1.38	10.2	1690	7.1	911	408	0.97
	1	200	10	220	279	1.76	8.1	1525	3.67	544	417	1.45
	2	200	20	250 以上 66	270	1.83	7.7	1490	2.38	307	414	1.36
	3	175	2		185	2.86	12.0	1637	0.5	114	319	0.87

[0050] The notes of a table 3: *1 ... Glass transition point. It is N2 by the TMA method. It measures with the programming rate of 2 degrees C / min in an air current.

* 2 ... Coefficient of linear expansion. The value below a glass transition point.

* 3 ... JIS It is based on K-6911.

* 4 ... It is N2 by the TGA method. It measures with the programming rate of 10 degrees C / min in an air current.

* 5 ... A pressure cooker test, 121 degree-C/100RH/100hrs.

[0051] The resin constituent of each example is remarkably excellent in the thermal resistance of a hardened material, and a mechanical characteristic compared with the general-purpose epoxy resin constituent (orthochromatic cresol novolak system epoxy resin constituent of the example 3 of a comparison) so that clearly from a table 2 and a table 3. Moreover, as a copolymerization component, compared with diaryl bisphenol A (example 1 of a comparison), and bisphenol A diaryl ether (example 2 of a comparison), whenever [for shaping / stoving temperature] is low, cycle time is short, and a moldability is good, and there is little absorptivity of a hardened material.

[0052]

[Effect of the Invention] The resin constituent for closure of this invention is excellent in fabricating-operation nature, the hardened material has little absorptivity and its thermal resistance is also good.

[Translation done.]